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DETERMINATION OF THE DIFFUSION COEFFICIENTS
IN ALLOYS WITH SEVERAL PHASES

by

V. T. Borisov
V. M. Golikov
G. N. Dubinin

Izvestiya, Akademii Nauk, SSSR, Metallurgiya i Gornoe Delo,
No. 4, 147-152 (1964)

Translated from the Russian by Harry Brooks

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Along with such widely known forms of thermo-chemical processing of alloys as cementation, nitridation, and cyanidation, certain new processes have acquired wide application in recent years. These include such processes of diffusion saturation of the surface of alloys as saturation with metals (aluminizing, chromizing, molybdenumizing, etc.) and saturation with metalloids (boronizing and siliconizing).

The extensive industrial use being made of these processes is due to the high grade of physico-chemical properties acquired by products as a result of diffusion saturation of their surfaces by metals, metalloids, or their compounds. Diffusion saturation of the surface of alloys with metals and metalloids is not only an effective means but, in many cases, the only possible means for solving such modern technological problems as the protection of the surface of alloys from oxidation at high temperatures, increasing their durability and resistance to corrosion, providing special electrical and magnetic properties on the surface, etc.

Despite the availability of a fairly extensive literature devoted to the study of individual questions of diffusion saturation of the surfaces of alloys with metals and metalloids (see, for instance, the Literature Cited^{1,2,3,4,5}), much less attention has been paid to calculating the diffusion constants in chemico-thermal processing.

The most thorough investigations in this area are referenced^{1,6,7,11} in the Literature Cited*. At the same time, a study of the laws of the diffusion of various elements when they are used to saturate the surface of metals and alloys makes it possible not only to broaden our understanding of the nature of phase transformations taking place in alloys during their processing, but also makes it possible to compute correctly the production rate of the diffusion saturation of the surface of alloys by means of metals and metalloids. Such computations are of special interest in connection with the increased use, in production, of vapor, gas, and liquid methods of saturating the surface of alloys by metals and metalloids since they make it possible to intensify the saturation process.

The diffusion constants for alpha-phase chromium used for the chromizing of iron in a gas medium have already been computed by

*Given the framework of the present article, we are not citing here the extensive literature available on general questions of diffusion. This can be had, for instance, from the bibliography cited by Gertsriken and Dekhtyar⁸.

Hoar and Groom⁹ while the corresponding figures for a liquid medium are given by Campbell et al¹⁰. However, the authors of these papers in using the basic diffusion equation

$$\xi = 2\nu\sqrt{Dt}$$

(where ξ is the thickness of the diffusion layer; ν , a constant which depends on concentration and temperature; t , the duration of the diffusion) conditionally accepted the value of ν as being equal to 1. The computed values of D and Q then become approximately as follows:

$$D_{\alpha}^{1100^{\circ}} = 14.7 \cdot 10^{-10} \text{ cm}^2/\text{sec},$$

$$Q = 57 \text{ Kcal/g} \cdot \text{at} \quad (\text{Hoar and Groom}^9),$$

$$D_{\alpha}^{1100^{\circ}} = 25.5 \cdot 10^{-10} \text{ cm}^2/\text{sec},$$

$$Q = 58 \text{ Kcal/g} \cdot \text{at} \quad (\text{Campbell et al}^{10}).$$

Buckle¹¹, using a method he had worked out for computing the diffusion coefficient for a multiphase layer formed on the surface of a sample, utilized the data given by Galmiche¹² on the diffusion-chroming of iron in a gas medium, and computed the diffusion constant for the alpha- and gamma-phase of chromium, taking into account the concentration factor of the parameter of ν .

$$D_{\alpha}^{1100^{\circ}} = 50 \cdot 10^{-10} \text{ cm}^2/\text{sec},$$

$$D_{\gamma}^{1100^{\circ}} = 0.92 \cdot 10^{-10} \text{ cm}^2/\text{sec}.$$

The present article examines the method for determining the diffusion coefficient in cases when a multiphase diffusion layer is formed on the surface zone of the metal.

The calculation of the coefficient of diffusion was made by using as example the chromium-saturation of iron in a gas phase.

Figure 1 presents schematically the distribution of concentration in a sample when, in the process of diffusion, several different phases are formed, $k = 1, 2, \dots, n + 1$.

In determining the diffusion coefficients for various phases D_k in the form of a concentration curve, we shall assume that the coefficients are constants within the limits of their respective phase and that the concentration on the outer surface of sample z_0 is not dependent on time.

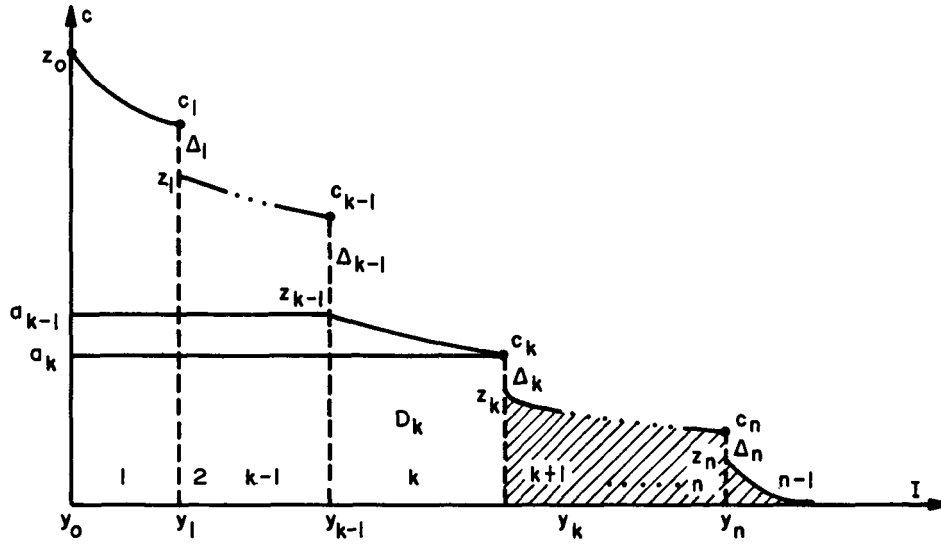


Figure 1. Schematic Distribution of Concentration of Diffusion in a Heterophase Medium

Concentrations z_{k-1} and c_k , on the phase boundaries, are also assumed to be known and not to be a function of time. These values can be determined in a diagram giving the state of the respective system if one assumes that the shift of phase boundaries occurs quasi-statically. Generally, the values of c_k , z_k , and values of the jumps or discontinuities $\Delta_k = c_k - z_k$ are determined experimentally. Thus, the concentration $c^k(x, t)$ within phase k satisfies the following equations:

$$\left. \begin{aligned} D_k c_{xx}^k(x, t) &= c_t^k(x, t) \\ y_{k-1}(t) &\leq x \leq y_k(t) \\ c^k[y_{k-1}(t), t] &= z_{k-1} \\ c^k[y_k(t), t] &= c_k \end{aligned} \right\} \quad (1)$$

for which the following expression is the solution

$$c^k(x, t) = A_k \operatorname{erf}(x/2\sqrt{D_k t}) + B_k \quad (2)$$

where

$$A_k = \frac{\delta_k}{\operatorname{erf}(b_{k-1}/\sqrt{D_k}) - \operatorname{erf}(b_k/\sqrt{D_k})},$$

$$B_k = \frac{c_k \operatorname{erf}(b_{k-1}/\sqrt{D_k}) - z_{k-1} \operatorname{erf}(b_k/\sqrt{D_k})}{\operatorname{erf}(b_{k-1}/\sqrt{D_k}) - \operatorname{erf}(b_k/\sqrt{D_k})},$$

$$\delta_k = z_{k-1} - c_k.$$

Within the limitations outlined above and in the presence of natural initial conditions $c^{n+1}(x, 0) = 0$, the boundaries separating the different phases shift—as we know—during the process of diffusion, according to the parabolic law.

It is, therefore, assumed in equations (1) and (2) that

$$y_k(t) = 2b_k\sqrt{t}. \quad (3)$$

The index k in equations (1) to (3) assumes the value of $1, 2, \dots, n+1$, with $y_0(t) = 0$ ($b_0 = 0$); while when $k = n+1$, we must regard $y_{n+1}(t) = \infty$ ($b_{n+1} = \infty$), $c_{n+1} = 0$ (see Figure 1). The mass balance on an arbitrary boundary, moving according to the law of $x = y(t)$, can be expressed by equation

$$dm/dt = -Dc_x[y(t), t] - c[y(t), t]dy(t)/dt \quad (4)$$

where dm/dt is the resulting flow across the phase boundary in question in the direction of an increase in the value of the coordinate x . The first and second terms on the right side of the above equation express, respectively, the pure diffusion flow and the flow resulting from the crossing of the phase boundary by the given substance because of the boundary shift. The quantity of matter m_k , which at a moment in time t is found beyond the border which separates phases k and $k+1$, moving in accordance with the law of $x = y_k(t) = 2b_k\sqrt{t}$, can be computed by the following two methods (numerically, the value of m_k equals the shaded area in Figure 1):

$$\begin{aligned} m_k &= -D_k \int_0^t c_x^k[y_k(t), t] dt - c_k y_k(t) \\ &= -D_{k+1} \int_0^t c_x^{k+1}[y_k(t), t] dt - Z_k y_k(t). \end{aligned} \quad (5)$$

The equivalence of the two methods is the result of the continuity of the flow across the phase borders.

Using equations (2) and (3), we can then compute the following:

$$\frac{m_k}{2\sqrt{t}} \sqrt{\frac{D_k}{\pi}} \delta_k \frac{\exp(-b_k^2/D_k)}{\operatorname{erf}(b_k/\sqrt{D_k}) - \operatorname{erf}(b_{k-1}/\sqrt{D_k})} - c_k b_k, \quad (6)$$

$$\frac{m_k}{2\sqrt{t}} = \sqrt{\frac{D_{k+1}}{\pi}} \delta_{k+1} \frac{\exp(-b_k^2/D_{k+1})}{\operatorname{erf}(b_{k+1}/\sqrt{D_{k+1}}) - \operatorname{erf}(b_k/\sqrt{D_{k+1}})} - z_k b_k. \quad (7)$$

By replacing k , in the last equation above, with $k - 1$, we get

$$\frac{m_{k-1}/2\sqrt{t} + z_{k-1}b_{k-1}}{m_k/2\sqrt{t} + c_k b_k} = \exp\left(\frac{b_k^2 - b_{k-1}^2}{D_k}\right) \quad (8)$$

which, solved for D_k , becomes

$$4D_k t = (y_k^2 - y_{k-1}^2) / \ln\left(\frac{m_{k-1} + z_{k-1}y_{k-1}}{m_k + c_k y_k}\right). \quad (9)$$

This expression can be used for determining the coefficient of diffusion in various phases. To find the coefficient D_k , it is sufficient to know the position of a given phase boundary at any given point in time t , and the relationship of the integral diffusion flows across these boundaries as of that particular moment, $\theta_k = (m_{k+1} + z_{k-1}y_{k-1})/(m_k + c_k y_k)$.

According to Figure 1, the value of θ_k represents the relationship between the areas of sections of the diffusion zone lying below the a_{k-1} line and the a_k line, respectively. It can be calculated directly if the distribution concentration in the given sample has been determined experimentally.

Formula (9), which for an area adjacent to the surface of the sample ($k = 1$) takes the form of

$$4D_1 t = y_1^2 / \ln\left(\frac{m_0}{m_1 + c_1 y_1}\right), \quad (10)$$

can be utilized for determining the coefficient of diffusion in all zones except the last one, since when $k = n + 1$ (i.e., when $y_{n+1} \rightarrow \infty$ and $c_{n+1} \rightarrow 0$), expression (9) becomes identical. Formula (7) can be used to determine the value of D_{n+1} . With $k = n$, it gives

$$\sqrt{\pi} \xi e^{\xi^2} \operatorname{erfc} \xi = \frac{z_n y_n}{m_n + z_n y_n},$$

$$\xi = \frac{y_n}{2\sqrt{D_{n+1} t}}. \quad (11)$$

The right side of this equation can also be determined graphically, if the course of the concentration curve in the last zone is known. The

function $\sqrt{\pi} x \exp(x^2) \operatorname{erfc} x$ is a monotone function, the value of which increases and tends to approach 1 for increasing values of x .

Following is a table of the values of this function, which can be used to find the required value of ξ in equation (11):

x	$\sqrt{\pi} \cdot x e^{x^2} \operatorname{erfc} x$	x	$\sqrt{\pi} \cdot x e^{x^2} \operatorname{erfc} x$	x	$\sqrt{\pi} \cdot x e^{x^2} \operatorname{erfc} x$	x	$\sqrt{\pi} \cdot x e^{x^2} \operatorname{erfc} x$
0.1	0.159	0.6	0.603	1.2	0.804	2.2	0.917
0.2	0.286	0.7	0.652	1.4	0.839	2.4	0.929
0.3	0.390	0.8	0.693	1.6	0.866	2.6	0.936
0.4	0.475	0.9	0.720	1.8	0.887	2.8	0.944
0.5	0.545	1.0	0.757	2.0	0.904	3.0	0.950

Expressions (6) and (7) can serve as a basis for another method of determining the diffusion coefficient of a multiphase medium. This method—unlike the one described above—does not require knowledge of the exact progress of concentration in the sample under investigation. Since expressions (6) and (7) are equal, we obtain the following recurrent equation

$$\delta_{k+1} \frac{2 \sqrt{D_{k+1} t}}{y_k} \cdot \frac{\exp(-y_k^2/4D_{k+1}t)}{\operatorname{erf}(y_{k+1}/2\sqrt{D_{k+1}t}) - \operatorname{erf}(y_k/2\sqrt{D_{k+1}t})} = \delta_k \frac{2 \sqrt{D_k t}}{y_k} \cdot \frac{\exp(-y_k^2/4D_k t)}{\operatorname{erf}(y_k/2\sqrt{D_k t}) - \operatorname{erf}(y_{k-1}/2\sqrt{D_k t})} - \Delta_k, \quad (12)$$

which makes it possible to determine the value of D_{k+1} if D_k has already been determined and if the position of the phase-boundaries at the time-instant t —as well as the concentration level at these points—is known. Thus, in order to find all the coefficients, it is sufficient—besides establishing the value of δ_k , Δ_k , and y_k , already mentioned—to know just the value of D_1 .

To find D_1 , convenient use can be made of the formula which determines the total mass m_0 of the substance which had diffused its way into the body of the sample. We must bear in mind, however, that in a number of cases it is possible to determine m_0 without defining the form of the concentration curve (e. g., by means of weighing the sample if saturation took place in a gas or liquid phase, or by measuring the total γ -radioactivity of the sample with the use of radioactive isotopes).

The equation for m_0 derived from formula (7) for the case when $k = 0$ has this form:

$$m_0 = 2\sqrt{D_1 t} \delta_1 / \sqrt{\pi} \operatorname{erf}(y_1/2 \sqrt{D_1 t}) . \quad (13)$$

By using formula (12), the coefficients D_2, D_3, \dots, D_{n+1} can be computed by the graphic method if we first draw up a family of functions $\exp(-x^2)/x(\operatorname{erf} \alpha x - \operatorname{erf} x)$ to determine the values of x and α at the intervals required ($x = y_{k+1}/2\sqrt{D_{k+1}t}$, $\alpha = y_{k+1}/y_k > 1$ for the left half of the equation, and $x = y_k/2\sqrt{D_k t}$, $\alpha = y_{k-1}/y_k < 1$ for the right).

By using the first of the methods described above, we were able, with the aid of formulas (9) to (11), to determine the coefficients for the diffusion of chromium in iron (with a 0.03% C content), for α and γ phases, saturating the iron with chromium in the gas phase¹³.

Figure 2, below, shows the results of determining the concentration of chromium by layers in the diffusion zone at temperatures of 900°, 1000°, 1100°, and 1200°C—the duration of the process being 3 and 6 hours.

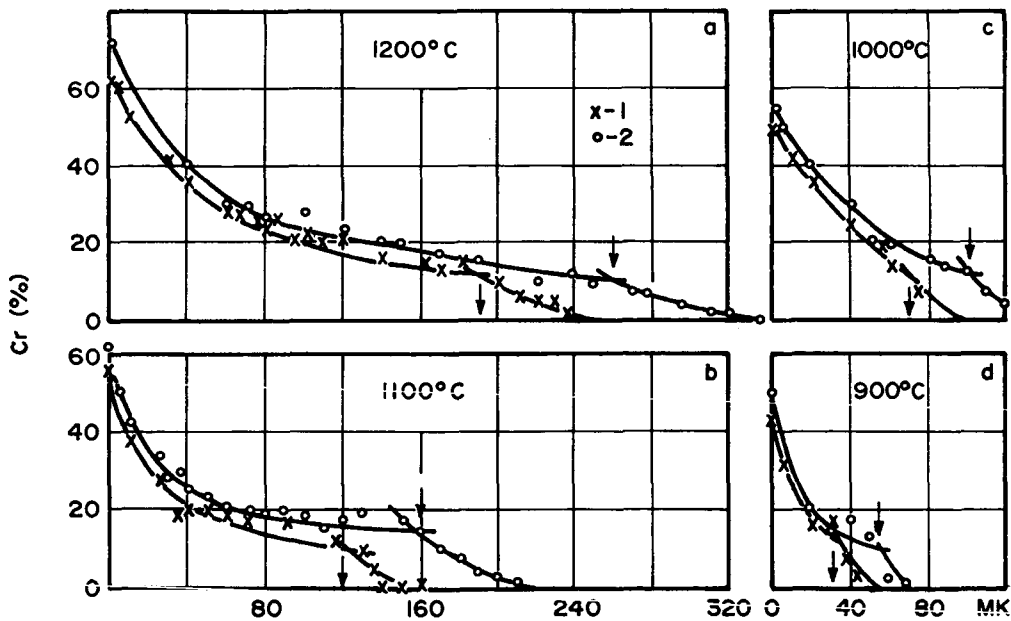


Figure 2. Changes in the Concentration of Chromium in the Surface Zones of Iron Which Underwent a Chromium Saturation Process in a Gas Medium at Temperatures from 900° to 1200°C for a Period of Three Hours (1) and Six Hours (2)

Chromium concentration by layers was determined by the spectral method¹⁴. The nodal points of the concentration curves (identified by

arrows in Figure 2) correspond to the boundaries of $\gamma \rightarrow \alpha$ phase-transition at an isothermic point in the process. The upper part of the curve corresponds to chromium distribution in the α -phase, while the lower part corresponds to the γ -phase (at annealing temperature). Chromium concentration on the boundary of the α and γ phases, obtained as a result of these experiments, corresponds satisfactorily to the results shown in the diagram of the Fe = Cr system state¹⁵.

The results of computing the chromium diffusion coefficient in the α and γ phases are shown in Figure 3, below.

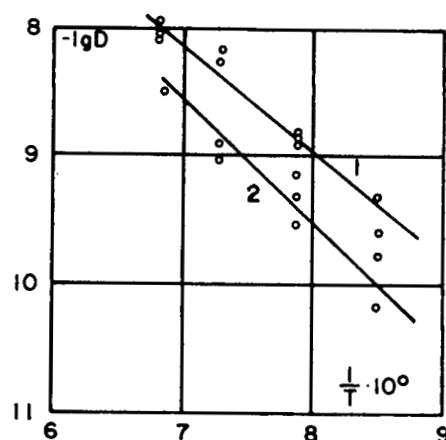


Figure 3. The Temperature Factor in the Coefficient of Diffusion (D , cm^2/sec) of Chromium in the α -State of Iron (Curve 1) and the γ -State (Curve 2)

Processing the experimental data makes it possible to present equations for the chromium diffusion coefficient in the α and γ phases in iron as follows:

$$D_{\alpha} = 1.55 \cdot 10^{-3} \exp\left(\frac{36,000}{RT}\right), \quad (14)$$

$$D_{\gamma} = 10^{-2} \exp\left(-\frac{44,000}{RT}\right). \quad (15)$$

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Spectral method						

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